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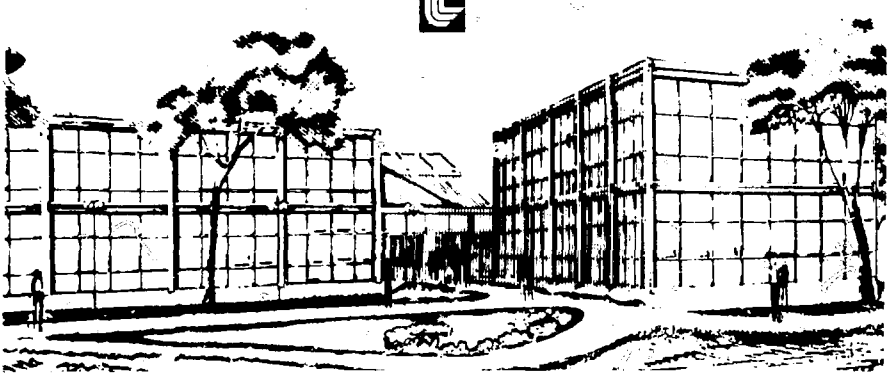

PROPERTIES OF NEUTRON-RICH NUCLEI STUDIED BY FISSION PRODUCT NUCLEAR CHEMISTRY

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INTRODUCTION

Accessibility to trends of nuclear properties over large ranges of mass number and constant proton or neutron number is an important factor in the development of modern theories. These properties must be correctly predicted for the lightest as well as the heaviest nuclei of a series. In addition, global models must be tested against reliable and detailed data from all regions of the nuclear chart. Understanding the cause of deviations of model predictions from experimental data often suggests important improvements to these models. In deed, it is considered by some that "one of the greatest tragedies of life is the murder of beautiful theories by ugly experimental facts" (Struble and Meyer, 1972). Currently there are numeous accesses to study the proton-rich (neutron-poor) nuclei. However, there is but one way to obtain far-from-stability neutron-rich nuclei: fission. The activity in this area of research has recently been reviewed at the "Workshop on the Spectroscopy of Fission Product Nuclei" (von Egidy 1979). In addition to extending the range of systematics, the exploration of fission-product nuclei also allows the exploration of different phenomena not accessible in proton-rich nuclei.

The utility of information derived from the study of fission products ranges from the quest of understanding our origins to problems associated with the design and monitoring of today's energy sources. Certainly input into cosmology problems of the R-process requires accurate base information derived from fission product data on masses, half lives, and nuclear structure (Schramm 1977, Takahshi 1977). The design of reactor cooling systems requires not only the measurement of decay heat by integral methods (Dickens 1978) but more advanced designs require summation calculations based on information concerning the individual contributor's decay properties (Reich

1977, Prussin 1974). The investigation of the fission yields by spectroscopy techniques requires fission product decay metrology (Denschlag 1978). In pursuing the acquisition of this fission product metrology we simultaneously are acquiring a base of information which allows us to explore fundamental properties of nuclei under conditions of high-excitation energy and large-neutron neutron excess. In turn some of the longer-range applied problems can only be solved by obtaining answers to fundamental questions such as the nature of beta-delayed neutron emitters including their energy spectra, daughter level population ratios and the related question of beta strength (Kratz 1978, 1979; Reich 1977).

Two techniques are employed in the isolation of fission products. One centers around separations dependent upon nuclear charge (Z-separations) and the other separates by mass (A-separation). The latter is performed by ISOL systems as will be described later in this symposium by Prof. Brenner (1979) and Prof. Hansen (1979). The use of Z-separators has been known in the past as CHEMISTRY. Its use for fission-product research is covered in detail elsewhere (Meyer 1979). After a short description of the techniques we use for elemental isolation we will discuss some recent results in the area of structure of nuclei near ^{132}Sn (Sec. 3.1), issues that impact on particle emission (Sec. 3.2), and coexisting structures in nuclei (Sec. 3.3).

2. TECHNIQUES USED IN ELEMENTAL ISOLATION

The techniques for chemical separation of fission products can be broken into two types; that of batchwize separation and that of continuous initial separation. However, one ought to recognize at the outset that many "continuous" techniques in actual fact produce the final measurable incremental (batchwize) amount, and because of effects such as source holdup times are not necessarily instantaneous. The decision as to whether to

employ batchwize or continuous conditions can often depend upon the problem to be attacked and the source of fission products available. However, when compared, the efficiency of batchwize operation relative to continuous operation generally shows that activities with half lives down to a few seconds can be studied by batchwize operation (Stevenson 1977, Meyer 1979).

For experiments using continuous separation techniques the fission products are removed from the fissioning site by either liquid flow through a target chamber or by use of a gas-jet recoil system (Trautmann 1975, Silva 1977). The latter designs use clusters in a carrier gas and are based on concepts discussed elsewhere (MacFarlane 1974). As diagrammed in Fig. 2.1, the gas jet is mixed with an aqueous phase, sent to a degassing unit, and then on to the first stage of a SISAK unit. The aqueous phase is mixed with an organic phase containing extractants. The rapid-phase separation occurs at the H-10 centrifuge where the mixed phases enter through a side port as shown at (A) in Fig. 2.2 and arrive at the inlet chamber (B) where it is accelerated to the rotational speed of the inner bowl. Upon acceleration the mixed phases are forced into a separation chamber (C) containing several isolated chambers symmetrically arranged about the central shaft. The separation chamber baffles (B) are slanted such that the heavier phase, forced outward by the centrifugal force is also forced downward toward the heavy-phase collecting chamber (E). Here it encounters a stationary spiral wheel (F) that, in concert with the rotational speed of the liquid, acts as a pump and forces the liquid up and out the heavy-phase outlet (G). Conversely, the light-phase path is through the upper chamber (H) to a light-phase pump (I) then up and out a light-phase exit (J). As diagrammed in Fig. 2.1 the centrifuges are run in series with mixers to perform initial extraction, selective extraction (III), and final back extraction cleanup (IV). The final centrifuge

is used to perform cleanup of the extractant before it is recycled. Experiments can, of course, use one up to several mixer-centrifuge stages.

The alternate technique to continuous separation is to perform chemical isolation of discrete amounts of gross fission products. This batchwize separation can be repreated a number of times until sufficient data is accumulated. The full cycle of operation for our automated system, shown in Fig. 2.3, starts with rabbits containing doubly-encapsulated samples of U-235 in solution being shot into the reactor from an automatic rabbit launcher. A computer controls the loading and firing from a magazine that holds 38 rabbits. A microprocessor loads and sends the rabbit through a pneumatic tube (activated by nitrogen gas) to the core of the Livermore Pool-Type Reactor (LPTR). The sample is irradiated for a specific time, then the microprocessor sends it through a second pneumatic tube to a receiver located in a chemistry laboratory hood approximately 45 m from the core of the LPTR. Upon arrival at the laboratory the rabbit is impaled by and punctured on a doubly concentric needle that extracts the fission product, adding acid in the process. Purging with N_2 gas cleans out the gases produced during fission and precludes their daughter activities from being observed in subsequent spectroscopy measurements. The actual chemical separation of an element such as Sb from the other fission products is made upon addition of sodium borohydride ($NaBH_4$) to the acidified fission products and carrier. The stibene and arsine (gaseous SbH_3 and AsH_3) is transported through a $CaSO_4$ trap that eliminates other hydrides produced by $NaBH_4$ reaction. The final step is destruction of the stibene with KOH in ethanol; the homologue AsH_3 passes through. This final step leaves the Sb on a substrate in front of a counting station. As soon as the final chemical step is finished, the microprocessor turns on the spectrometers for measurement. An alternate procedure uses a $CaSO_4$/NaOH trap for the study of short-lived As isotopes.

3. CURRENT RESEARCH

3.1 Structure of Nuclei Near ^{132}Sn

It has become possible to circumvent many of the problems attendant with performing microscopic calculations in the ^{132}Sn region. This is through the application of numerical methods of diagonalizing very large vector space with the Lanczos algorithm (Lane 1979). In order to test and improve these calculations we have been investigating the properties of ^{132}Te, ^{133}Te, and ^{134}Te through Rapid Automated Nuclear Chemistry (RANC) studies of the ^{132g}Sb, $^{132}Sb^{m}$, ^{133}Sb, and ^{134}Sb decay. These studies have been performed over a period of several years and include singles, $\gamma\gamma$-coincidence, $\gamma\gamma t$-coincidence, and multiscale spectroscopy using the LLL autobatch facility. Studies of the shorter-lived 10 s ^{134}Sb decay were performed on our improved facility. All but the 10 s ^{134}Sb experiment used an ion exchange resin to concentrate the antimony for measurement. The 10 s ^{134}Sb required a different catcher because the kinetics of ion exchange were too show to be effective. Coincidence measurements for the latter required between 10 and 15 working days to acquire the requisite 25 x 10^6 events for proper evaluation.

The decay of the ^{132}Sb isomers provide a near-complete description of the ^{132}Te levels and their properties at excitations below 3 MeV. This includes the population of a 3.9 μs isomer proposed to have $J^{\pi} = 10^{+}$ by Sistemich (1979) and an 8^{+} level at 2701 keV. Some evidence for a lifetime of approximately 50 ns for the latter level can be understood in that the level arises predominately from the two-neutron hole states while the only levels accessible to its deexcitation are nearly pure proton particle excitations. We show a comparison of our experimental and theoretical results in Fig. 3.1.1. These and other properties are discussed elsewhere (Lane 1979, Henry 1979, Meyer 1979).

Studies of the mass 133 fission product chain has revealed a number of interesting features of these far-from-stability but near-closed shell nuclei. These include properties of the two-particle one-hole (2p1h) nucleus ^{133}Te populated in ^{133}Sb decay; near-closed shell test of the cluster vibration model in ^{133}I populated in the decay of ^{133}Te^{g} and ^{133}Te^{m} (see Fig. 3.1.2); the surprisingly wide variation in the values for the unique first forbidden beta decay for $7/2^{+} \rightarrow 11/2^{-}$ beta transitions with log f_1t values from greater than ten down to the fastest known of all β decays at 8.51 for ^{133}Sb decay as shown in Fig. 3.1.3 (Denschlag 1979, Meyer 1979); and the unexpected population of high-spin isomers via γ-ray cascades of increasing angular momentum following beta decay to high-energy levels (Griffin 1979).

We have applied the method of Whitehead (1974, 1976) as expanded by Hausman (1977) to nuclei with up to 5 excitons in the ^{132}Sn region. The potential well in which the valence particles moved was approximated by a spherical harmonic oscillator well matched in size. The interaction between valence nucleons was one due to Petrovich, McManus, and Madsen (Petrovich 1969). This force, which is a modified form of the realistic Kalio-Kolltveit interaction (Kallio 1964), has been used successfully for structure calculations in other regions of spherical nuclei. The single-particle energies were obtained from the ^{132}Sn one-neutron-hole spectrum (the $1h_{11/2}$ energy was readjusted in a few cases). In Fig. 3.2.4 we show the level of ^{133}Te up to 2200 keV which we observe in the decay of ^{133}Sb. These levels are compared to those we calculate in Fig. 3.2.5. When it is kept in mind that these calculations were made without the freedom of adjustable parameters, the agreement with experiment is found to be surprisingly good. We find the first three levels (J^{π} of $3/2^{+}$, $1/2^{+}$, and $11/2^{-}$) to be approximately 80% pure single-hole states in agreement with neutron pickup experiments. Below 2000 keV only

the remaining $3/2^+$ and the $5/2^+$ positive parity levels are fragmented while all but one $11/2^+$ level (at 1611) is predicted to be on the order of 80% pure being constructed from $|\pi g_{7/2}^2\ \nu d_{3/2}^{-1}\rangle$ or $|\pi g_{7/2}^2 \nu d_{5/2}^{-1}\rangle$ configurations. The 1496 $5/2^+$ level is mainly a mixture of $|\pi g_{7/2}^2\ \nu d_{3/2}^{-1}\rangle$ and and $|\pi g_{7/2}^2\ \nu d_{5/2}^{-1}\rangle$ (Lane 1979).

3.2 Particle Emission

A second example of work being done using chemical isolation systems centers around issues dealing with processes at high-excitation energies. The measurement of neutron spectra with high-resolution ^{3}He ionization chambers has led to the discovery of spectra with pronounced peak structure that have spacings that are large when compared to level density calculations (Franz 1974, Kratz 1978). The conclusions concerning the spectral shape and beta-intensity distribution are quite different from those arrived at by a purely statistical model approach (Hardy 1978, Hansen 1974). More detailed statistical model calculations (Prussin 1979) show that it is not possible to obtain simultaneous agreement between the general shape of the neutron-energy spectra and the experimental data on neutron-branching ratios to individual levels in the daughter nucleus such as populated in the $^{85}As \xrightarrow{\beta^-} {}^{85}Se^* \xrightarrow{n} {}^{84}Se$ sequence. Thus, a very important component in the solution of this problem is a precise knowledge of the ^{84}Se level structure available to the beta-delayed neutron emission of ^{85}Se*. Only five levels at 1455, 2122, 2700, 3541, and 5159 keV were known when investigations into the decay of ^{84}As to levels of ^{84}Se were undertaken.

We studied the decay of 5 s ^{84}As by simultaneous measurement of high-energy multiscaled singles and three-parameter ($\gamma\gamma t$) coincidence spectroscopy. The total chemistry time using the LLL autobatch system was 1.8 s. Approximately 10,000 separate separations were required to accumulate 25 million

coincidence events in approximately 10 working days. This allowed the identification of 28 levels below 4.75 MeV and the evidence of levels up to 9.5 MeV in ^{84}Se. These levels are in agreement with other N=50 nuclei and similar to the N=34, Z=28 nucleus.

3.3 Coexisting Structure in Both Neutron Poor and Neutron Rich Nuclei

Recently we have performed unified-model calculation for the description of the odd-mass in isotopes without introducing rotational states of the particle-core (Cd) coupled configurations as unspecified states. All single-hole ($1g_{9/2}^{-1}$, $2p_{1/2}^{-1}$, $2p_{3/2}^{-1}$, $1f_{5/2}^{-1}$) and 1p-2h states [seniority $v = 1$ and $v = 3$ with $p\epsilon(2d_{5/2}, 1g_{7/2}, 3s_{1/2}, 2d_{3/2}, 1h_{11/2})$] were considered, together with the collective excitations of the underlying Sn core (quadrupole as well as octupole phonons). The calculation was performed in a two-step procedure: First, by treating the Cd nuclei in a hole-core (Sn) coupling calculation. Afterward, we treated simultaneously the hole-core (Sn) and particle-core (Cd) configurations and therein showed the precise structure of the Cd-core states and their relations with the hole-core (Sn) configurations (Heyde et al. 1978). In our calculations, both the vibrational multiplet $|1g_{9/2}^{-1}Sn(2_1^+J); J^\pi\rangle$ and the rotational-like sequence $J^\pi = 1/2_1^+$, $3/2_1^+$,... of positive-parity states at low-excitation energy ($E_x \simeq 1.0$ MeV) result and are seen to interact only weakly. One exception is the $J^\pi = 5/2^+$ levels, where moderate mixing occurs. In the calculation of spectroscopic factors for the ^{114}Cd(^{3}He,d)^{115}In reaction our results equal those from a Nilsson-model calculation in which the positive-parity levels arise from a rotational band on top of the $1/2^+[431]$ orbital. The unified model is able to explain, by means of the 1p-2h excitations, the more phenomenological approach, where proton single-particle excitations through the Z=50 closed shell are coupled to the vibrational excitations of the Cd

core nucleus. The negative quadrupole moments for the rotational-like sequence of levels are also reproduced in the Unified-model approach. The reduced E2 transition probabilities B(E2) inside this rotational-like sequence stongly suggest a purely rotational explanation. In addition, we have presented evidence for similar-type excitations in Z=47 Ag nuclei (Glascock 1977).

These same type excitations are now being observed in the studies of proton-rich Z=79 and 81 nuclei by the UNISOR collaboration (Wood 1979). However, until now no evidence has evinced for their occurrence in odd-neutron nuclei. Some evidence for these type excitations in N=49 nuclei is emerging from RANC studies of the short lived arsenic fission products. The ^{83}Se levels at 963, 1062, and 2076 keV that are populated in the decay of 13s ^{83}As also have been shown to have strong $d_{5/2}$ particle strength in the ^{82}Se(d,p)^{83}Se reaction studies (Lin 1965, Montestruque 1979). These levels with J^{π} values of $3/2^+$, $1/2^+$, and $5/2^+$, respectively, may represent the first evidence for the occurrence of rotation-like bands in the N<50 odd-mass nuclei. Further investigation into the properties of these levels of ^{83}Se as well as similar levels identified in the lighter odd mass Se nuclei are necessary before detailed tests can be made.

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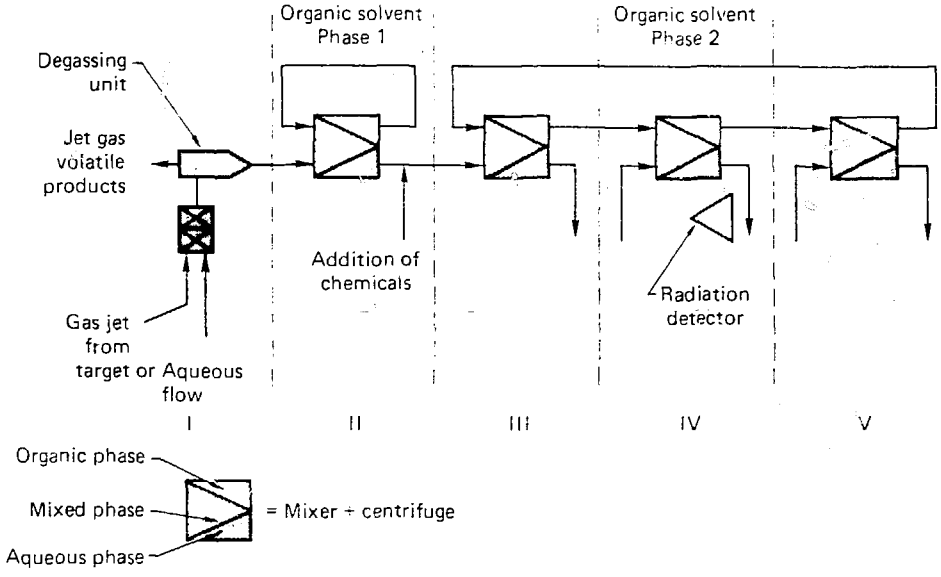


Fig. 2.1 Flow diagram for a SISAK based continuous chemistry experiment.

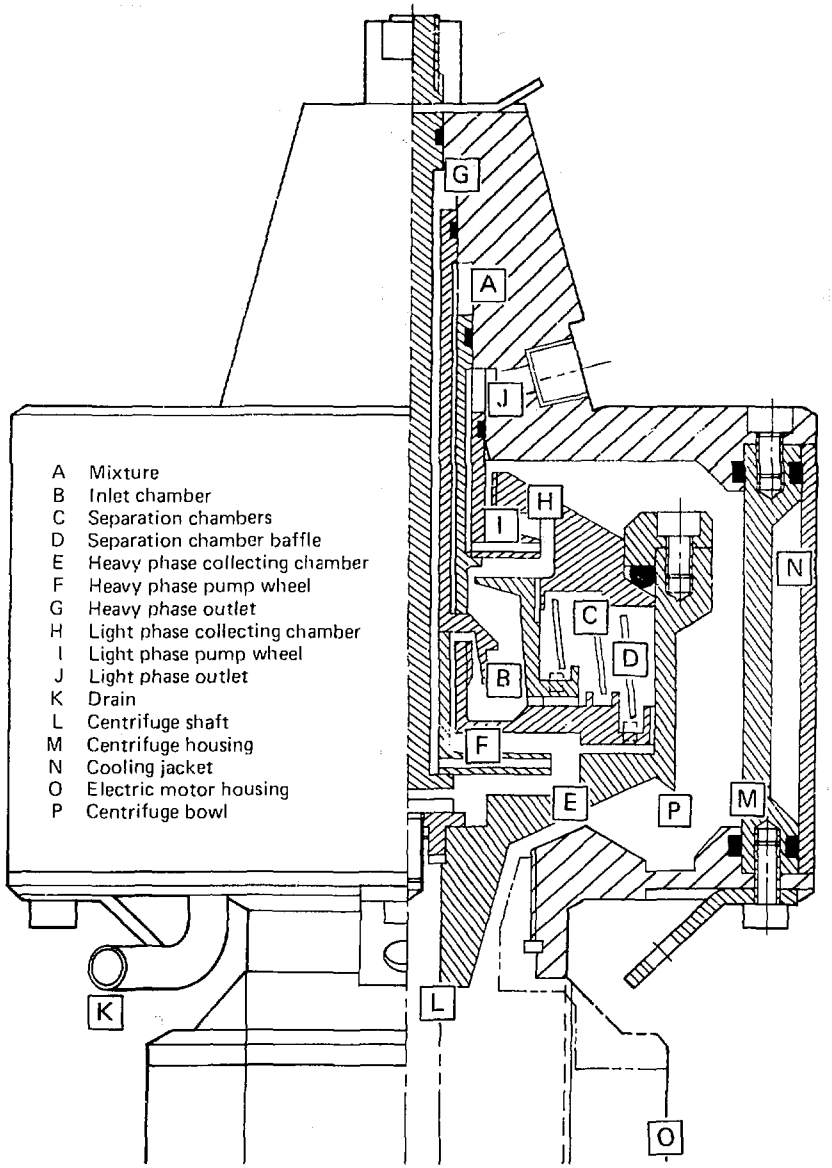


Fig. 2.2 Internal parts of an H-10 centrifuge (courtesy of Prof. J. Rydberg, Chalmers University and Dr. H. Reinhardt MEAB, Valnötsgatan 18, S-421 74 V. Frölunda, Sweden).

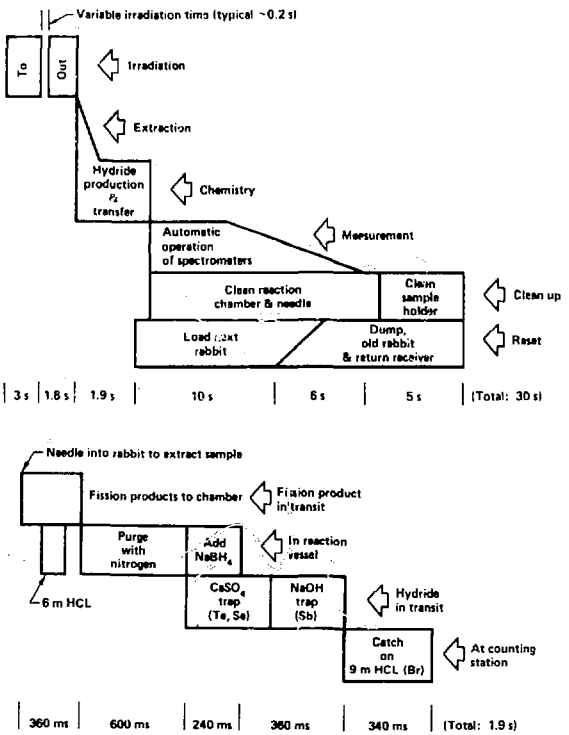


Fig. 2.3 Operation cycle of LLL autobatch facility - a) full time cycle, b) chemical separation sequence.

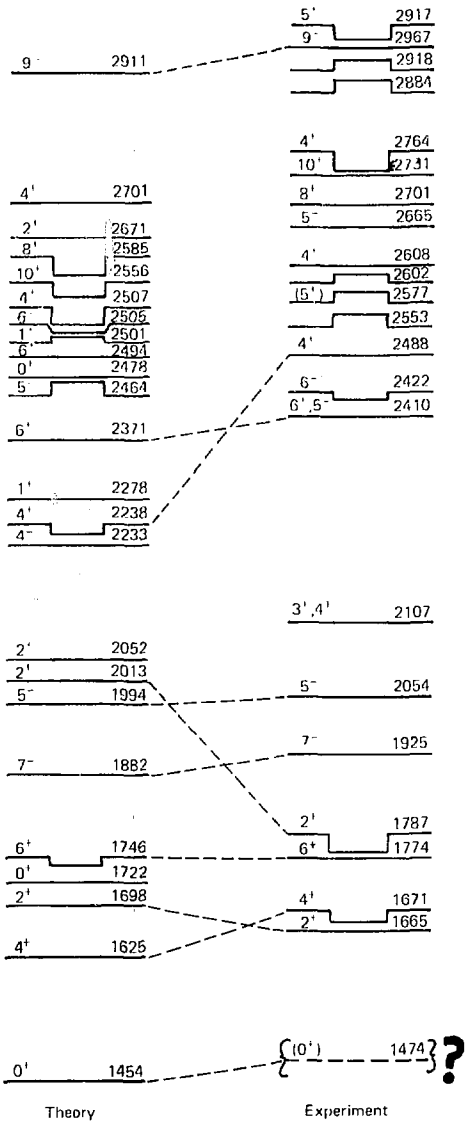


Fig. 3.1.1 Comparison of ^{132}Te levels from experimental results of the decay of the ^{132}Sb isomers and shell model calculations.

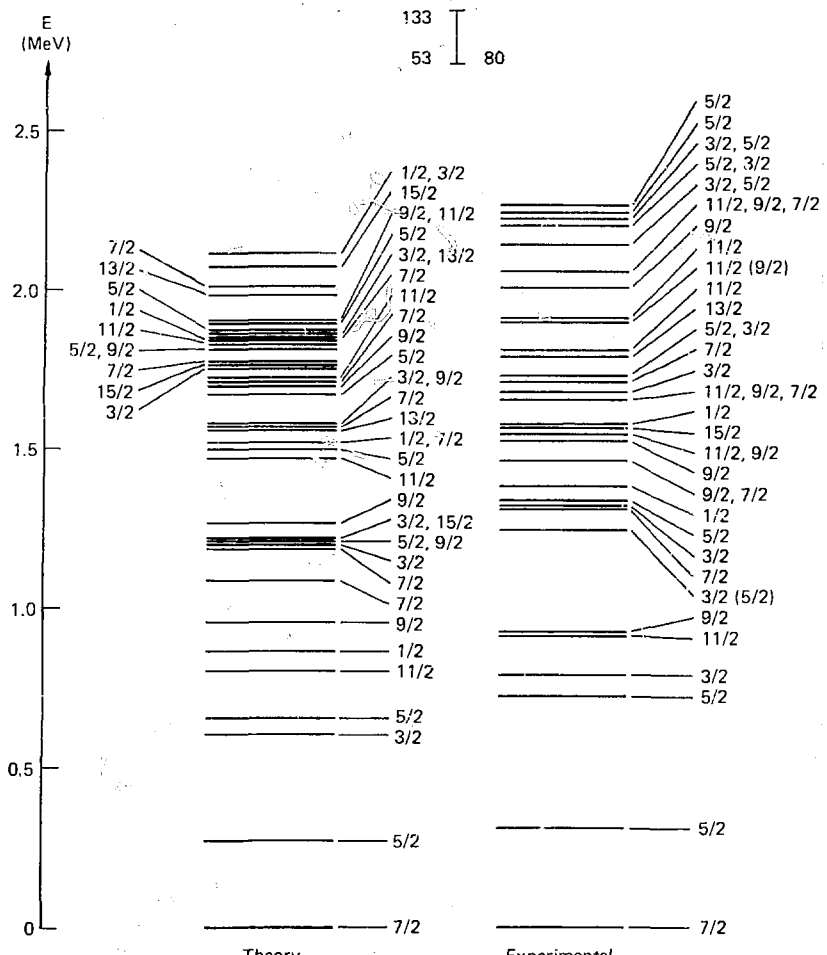


Fig. 3.1.2 Comparison of ^{133}I levels obtained by RANC studies of ^{133}Te isomer decays and cluster-vibration model calculations.

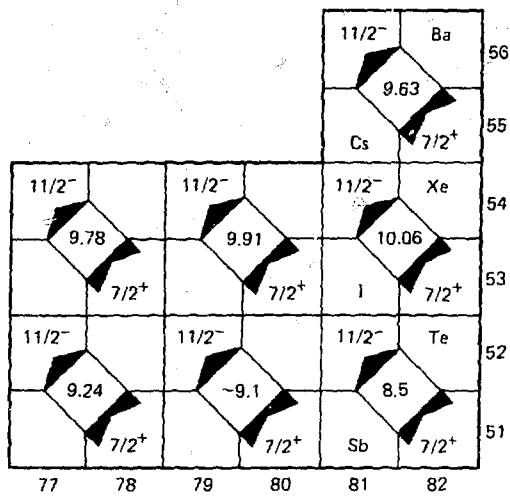


Fig. 3.1.3 The unique first forbidden beta decay log f_1t values for $7/2^+ \rightarrow 11/2^-$ transitions in the ^{132}Sn region.

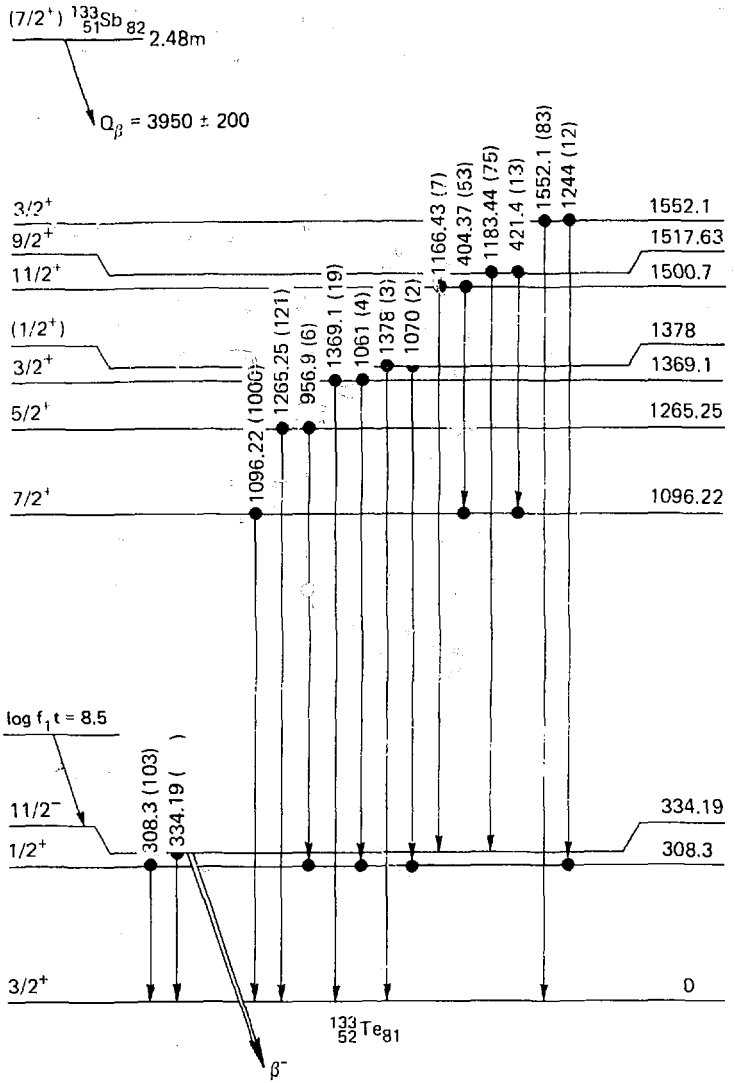
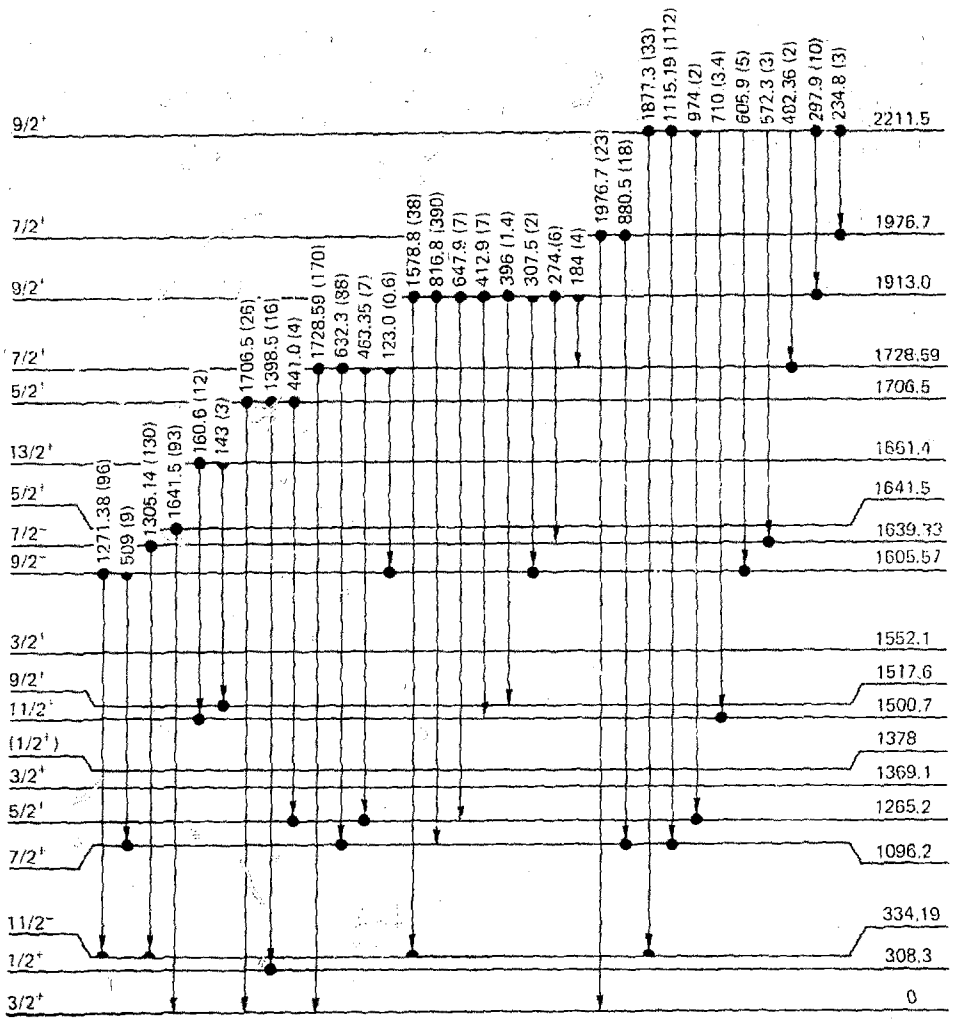


Fig. 3.1.4 Levels of ^{133}Te populated in the decay of ^{133}Sb
a) up to 1552 keV



$^{133}_{52}Te_{81}$

Fig. 3.1.4 Levels of ^{133}Te populated in the decay of ^{133}Sb - 1600 to 2250 keV.

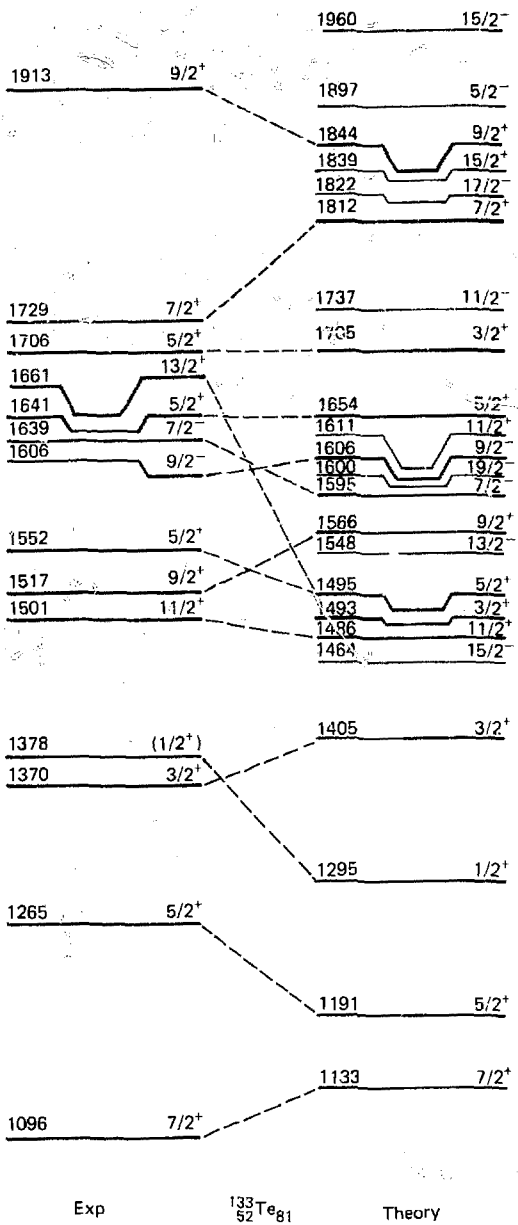


Fig. 3.1.5 Comparison of levels of ^{133}Te populated in RANC studies of ^{133}Sb decay and shell model calculations.